THE

ONTARIO WATER RESOURCES

COMMISSION

REPORT ON

A RECONNAISSANCE SURVEY

OF

RADIOLOGICAL POLLUTION

IN THE

SERPENT RIVER WATERSHED

BY

URANIUM MILL WASTES

1964

A RECONNAISSANCE SURVEY OF RADIOLOGICAL POLLUTION IN THE SERPENT RIVER WATERSHED BY URANIUM MILL WASTES

April, 1964

by

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C O N T E N T S

REPORT

ONTARIO WATER RESOURCES COMMISSION

Municipality: Improvement District of Elliot Lake. **Date of Inspection:** Oct. 3, 4, 1963.

Re: A Reconnaissance Survey of Radiological Pollution in the Serpent River Watershed by Uranium Mill Wastes.

Field Inspection by: J. H. Neil, C. F. Schenk, **Report by:** D. P. Caplice, J. H. Neil. G. J. Hopkins.

INTRODUCTION

Since 1957, the Ontario Water Resources Commission has been actively engaged in carrying out investigations in the Elliot Lake area directed primarily towards minimizing the chemical and physical effects of uranium mill wastes on the Serpent River watershed. This program has stressed the necessity of properly neutralizing all mill wastes followed by impoundment of these wastes in tailings ponds. With proper operation and maintenance of such ponds it is possible to greatly restrict discharges to surface waters, and in addition these ponds are usually successful in removing most settleable solids.

Despite this program, it is generally recognized that marked changes in the physical and chemical make-up of the streams and lakes in this watershed have occurred since the mines began operation. These have included: a change in colour of the lake waters that receive tailings decant directly; an increase in hardness, nitrate and sulphate concentrations of these waters; the deposition of tailings or other settleable solids in lakes used for impoundment areas, as well as a build-up of radionuclides which were formerly associated with the product recovered in mill operations.

Investigations into possible radioactive contamination were begun by the Ontario Department of Health in the summer of 1958. In 1961, increased analytical facilities were available and a monitoring program began, the main purpose of which was to watch for a build-up of radioactive materials in waters used for human consumption. It should be noted here that, during 1963, three

of the mines operating in the area were requested to find new sources of domestic water by the end of 1964, as it was becoming evident that levels of radium in the lakes from which water was being drawn for processing and drinking, were above tolerable limits.

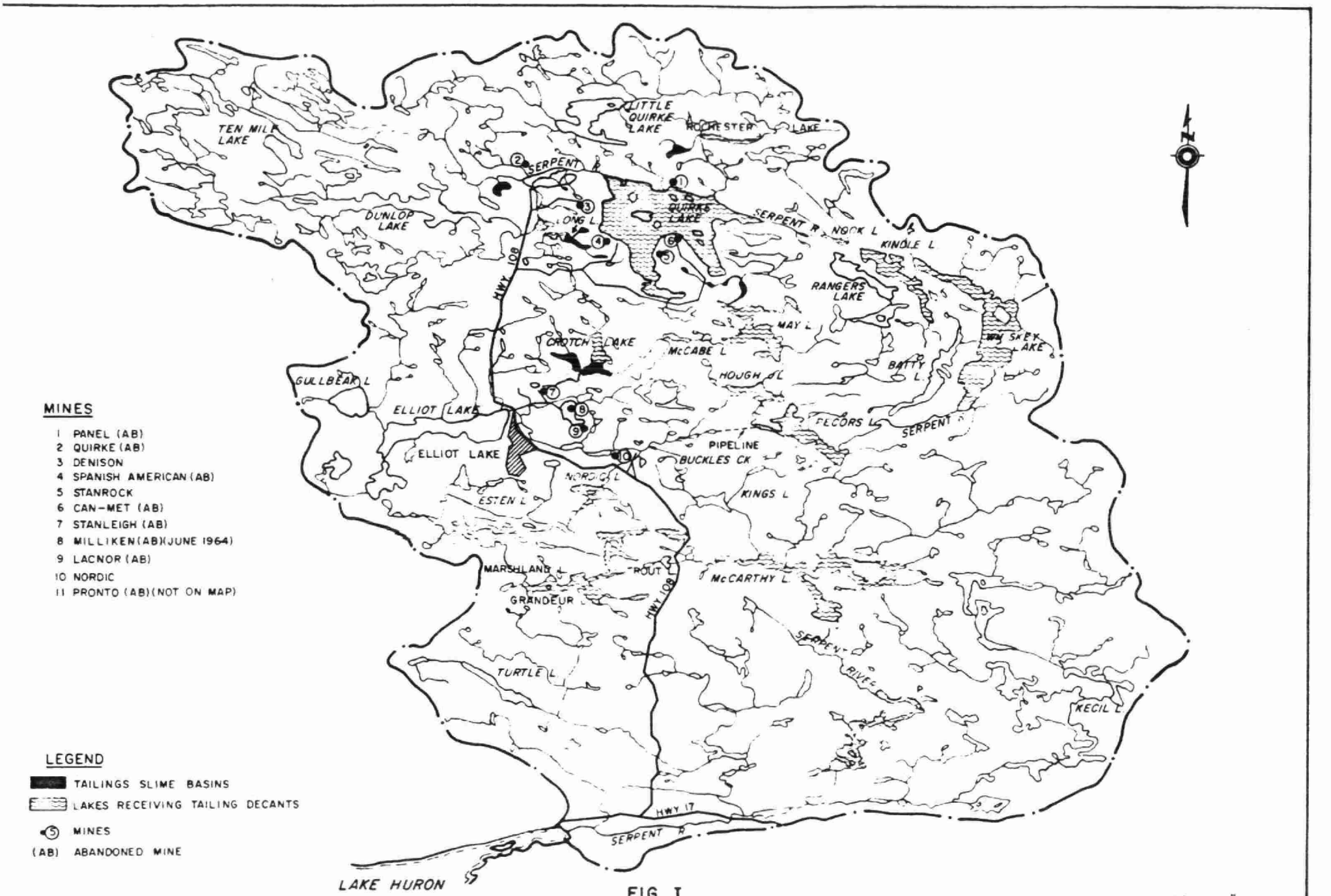
Radioactive substances are known to accumulate in living organisms. Various elements are taken passively from their environment while others are actively accumulated for skeletal and metabolic purposes. No sampling of living material for the purpose of determining the level of accumulated radioactive substances had previously been made in the Elliot Lake area and in view of the observed increase in the radioactivity of the water this seemed desirable.

The uranium mining area in the Algoma District of Ontario lies within the drainage basin of the Serpent River. (Fig. 1). This basin covers an area of 492 square miles and the river has an estimated mean annual flow of 615 cfs. The outflow of this system enters the north shore of Lake Huron near the Village of Spragge, 100 miles east of Sault Ste. Marie.

Intensive uranium mining operations began in the area in 1957, and at the peak period May, 1958, 11 mines were operating. Since that time, due to a severe outback in the world demand for uranium, the number of operating mines has been reduced to 4. The tonnages, names, start-up dates and shut-down dates for the 11 mills are shown in Table 1.

START-UP DATES, MILLS, TONNAGES AND SHUT-DOWN DATES

Tons per day	Mill	Start-Up		Shut-Down	
1,500	Pronto	September,	1955	April,	1960
3,000	Quirke	September,	1956	February,	1961
3,000	Nordic	January,	1957		
6,000	Denison	May,	1957		
4,000	Lacnor	September,	1957	June,	1960
3,000	Panel	February,	1958	June,	1961
2,000	Spanish American	May,	1958	February,	1959
3,000	Stanleigh	March,	1958	January,	1960
3,000	Stanrock	March,	1958		
3,000	Milliken	April,	1958	June,	1964
2,500	Can-Met	March,	1958	March,	1960


MINES
1 PANEL (AB)
2 QUIRKE (AB)
3 DENISON
4 SPANISH AMERICAN (AB)
5 STANROCK
6 CAN-MET (AB)
7 STANLEIGH (AB)
8 MILLIKEN (AB) (JUNE 1964)
9 LACNOR (AB)
10 NORDIC
11 PRONTO (AB) (NOT ON MAP)
LEGEND
TAILINGS SLIME BASINS
LAKES RECEIVING TAILING DECANTS
MINES
(AB) ABANDONED MINE
TEN MILE LAKE
LITTLE QUIRKE LAKE
ROCHESTER LAKE
SERPENT R.
DUNLOP LAKE
LONG L.
QUIRKE LAKE
NOOK L.
KINDLE L.
RANGERS LAKE
HWY 108
MAY L.
CROTCH LAKE
McCABE L.
HOUGH L.
BATTY L.
SKEY LAKE
GULLBEAR L.
ELLIOT LAKE
RECORS L.
PIPELINE
BUCKLES CK.
NORDIC L.
ESTEN L.
KINGS L.
MARSHLAND L.
TROUT
McCARTHY L.
GRANDEUR
TURTLE L.
KECIL L.
HWY 17
LAKE HURON
FIG. I
SCALE 1" = 4 MILES

All mines shown, except Pronto, were working, or are working, properties located in the upper portion of the Serpent River watershed located primarily in and around Quirke Lake. Production at mines in the Elliot Lake area is based on mining uranium-bearing ore, and concentrating the metal as uranium oxide U_3O_8.

The ore is crushed, ground, and leached with sulphuric acid to dissolve the uranium. The coarse sands are separated and discharged to waste as tailings, and the liquid portion is fed to ion exchange resin columns. Here the uranium is extracted from the feed solution, and is, in turn, stripped from the resin by nitric acid, precipitated, filtered, and dried to form yellow cake for shipping. The ion exchange feed solution having been stripped of its uranium is neutralized with lime and discharged with the tailings. From the start of mining operations until the end of 1963, 44,986 tons of U_3O_8 or 38,000 tons of uranium as the element have been mined in the Elliot Lake area.

SOURCES OF WASTE AND TREATMENT

The main sources of waste from the milling process are (1) the tailings, or finely divided waste rock that remain after the separation from the uranium-bearing acid solution and, (2) the barren solution that remains following the removal of the uranium oxide concentrate from the acid solution by ion exchange.

At all plants in the area these acidic wastes are neutralized with slaked lime and discharged as a slurry to a tailing disposal area.

A third waste at all mines consists of the mine water that is pumped from underground. In the early operational period of the mines, this water was discharged to either the tailings area or to the nearest watercourse, but in most cases it is now being used in the grinding circuit.

in the mill as it carries a significant concentration of uranium that can be recovered.

Coming now to the radioactive aspect of waste disposal from this industry, a brief outline of the problem and how it arises is called for.

Uranium^{238} is the first, or parent, of a long series of radioactive isotopes found in nature and known as the uranium-radium family of elements. Uranium^{235} is the parent of an entirely separate naturally-occurring series, the uranium-actinium family of elements. Uranium found under natural conditions consists of 99.28 per cent U^{238} and 0.71 per cent U^{235}. Hence, from the viewpoint of radioactive waste disposal, the uranium-radium family of elements is the one of major concern. U^{238} decays by emission of alpha and beta particles through (13) elements to a final product, lead (Pb^{206}), which is non-radioactive. Only the uranium is recovered from the ore, and all of the remaining members of the series are included in the process wastes.

The gross radioactivity of the mill waste material is attributable to each member of the uranium-radium series originally present in the ore. However, the relative degree of hazard presented by each of these isotopes covers a wide range with radium^{226} being the most hazardous, having the lowest Maximum Permissible Concentration in water of any of the 264 isotopes considered by the National Committee on Radiation Protection and the International Commission on Radiological Protection. The amount of radium^{226} contained in the ore delivered to uranium mills is very large relative to what are considered safe concentrations, and as essentially all of this radium will be contained in the mill wastes the fate of this one isotope in the rivers and lakes affected by mill wastes will be the primary topic of discussion in this report. Radium is known to be present in ore mined in the Elliot Lake area at a ratio of about $1 : 3 \times 10^{6}$. Using the known production of Uranium of 38,000 lbs., it may be calculated that about twenty-five lbs. (11,400 curies)

of pure radium have been discharged to tailings disposal areas.

Since the start of mining operations in the Elliot Lake area it has been stressed that all wastes should be retained in tailings ponds. This measure permits the settleable waste or solids containing the major portion of the radium to be confined, and, only in the cases where significant leaching of these wastes take place, should radium^{226} build-up in receiving waters.

The amount of overflow from a tailings pond is a function of net liquid input and the evaporation and seepage which occur. At the mines in the Elliot Lake area where actual lakes were used as tailings basins this net liquid input is much larger than would be encountered at a mill that had taken advantage of natural ground contours to create a tailings pond and presents a more difficult problem when methods of minimizing tailings pond overflows are considered. Of the ten mines that operated in the Elliot Lake area, five used lakes as tailings basins.

Dissolved radium concentrations in tailings pond waters can be expected to vary greatly depending upon such variables as differences in the raw ore, size of disposal pond, evaporation and mill solution bleed-off rates and co-precipitation of radium.

In summary, the beneficial effects of tailings ponds have been: (1) removal of settleable solids and some limited evaporation of liquids, (2) providing an opportunity for seepage which is desirable except where groundwater pollution is of significance and (3) retaining radioactive pollutants, temporarily permitting the decay of shorter-lived materials.

Undesirable features of such ponds have been: (1) the inherent porosity of the tailings permitting seepage laterally or vertically, (2) the prolonged contact between the liquid and the tailings, providing the opportunity for continued leaching of radioactive contaminants, and, (3)

their observed tendency to fail structurally and release their contents if the walls are not properly constructed and maintained.

The third feature of dyke failure is an important consideration at only two mines in the Elliot Lake area and appropriate measures should be taken in each case to preclude this occurrence. A protective secondary dyke surrounding the tailings pond area would, if properly designed and constructed, contain wastes accidentally released by tailings pond dyke failure. Such a dyke would also prevent the washing away of tailings by excessive surface water runoff.

UNITS OF RADIOACTIVITY

The amount of radioactivity present in a sample of any kind of matter, regardless of the element or elements which give rise to the activity, is stated in terms of the curie unit.

Originally the curie was considered to be the number of disintegrations occurring per second (d.p.s.) in one gram of pure radium. The modern definition is that one curie = 3.7×10^{10} d.p.s. for either pure or mixed isotopes. Since the curie represents such a large number of disintegrations per second, submultiples of this unit, namely millicuries, microcuries and micromicrocuries are used.

Unit	Symbol	Value
one curie		$= 3.7 \times 10^{10}$ d.p.s.
one millicurie		$= 3.7 \times 10^{7}$ d.p.s.
one microcurie	μc	$= 3.7 \times 10^{4}$ d.p.s.
*one micromicrocurie	$\mu\mu$c	$= 3.7 \times 10^{-2}$ d.p.s.
*one micromicrocurie		= one picocurie (pc)

The concentration of radioactivity in water or food is stated in terms of curies, or suitable submultiples, per unit volume. In this report radium concentrationsare stated as either micromicrocuries per litre for water or micromicrocuries per gram for solids. The Department of Health in Ontario used the term picocurie (pc) which is equivalent to micromicrocurie.

MAXIMUM PERMISSIBLE CONCENTRATIONS

Radioactive substances are to be regarded as poisons, some being more potent than others. Ingestion is to be avoided as far as possible, but complete avoidance is hardly feasible. There has always been some natural radioactivity in lakes and rivers and this appears to do no harm. Therefore, it is a question of attenuation, or at what concentration water containing a certain radioisotope is safe for normal recreational and domestic purposes, during a lifetime exposure.

In an attempt to answer this question, various authorities have put forward what are known as "maximum permissible concentrations" for specific radioisotopes. These values are rooted partly in experimental fact and partly in theory, and in some cases there is a lack of unanimity on the value to be used. Generally speaking, the standards are conservative: in deriving them it is assumed that the population exposed to water polluted to the MPC level will consume about 2 litres/day per person for fifty years, the requirement then being that no conceivable injury should result.

As noted above, radioisotopes vary in toxicity and MPC's vary accordingly. Mixtures of radioisotopes frequently occur in an environment and it is possible, and sometimes necessary, to set up an MPC for the mixture based on MPC's of the components. However, in some of these cases, one component stands out so strongly above the others that its MPC becomes

the controlling feature. This, it is felt, applies in the case of the Elliot Lake area where radium is the most significant radioactive nuclide found in the process wastes from the uranium mills. As previously mentioned, in terms of its MPC in domestic water supplies and food, radium is considered to be the most hazardous of all known radioisotopes. The OWRC in its publication "Drinking Water Objectives" has indicated an MPC for radium^{226} of 3.0 micromicrocuries per litre. As regards allowable concentrations of radioisotopes in food, the U. S. Atomic Energy Commission indicates that the values for specific radioisotopes used for water are also applicable to foods on a wet (live) weight basis. Thus the allowable concentration of radium in foods is 3.0 micromicrocuries per kilogram of live material.

The MPC values for members of the uranium-radium family of radioisotopes are shown in the Table II below:

T A B L E II

*URANIUM-RADIUM FAMILY, MPC_w VALUES[a]

Isotope	MPC_w μμc/l	Critioal Organ	Half-life		Emmission
Ra^{226}	3.3	Bone	1,620	yr.	Alpha
Pb^{210}	33	Kidney	22	yr.	Beta
Po^{210}	233	Spleen	140	days	Alpha
Th^{230}	667	Bone	8×10^4	yr.	Alpha
Th^{234}	6,667	GI tract	24.1	days	Beta
U^{234}	10,000	GI tract	2.5×10^5	yr.	Alpha
U^{238}	13,300	GI tract	4.5×10^9	yr.	Alpha

TABLE II continued

Isotope	MPC_w μμc/l	Critical Organ	Half-life		Emission
Bi^{210}	13,300	GI tract	5	days	Beta
Pa^{234}	b	-	1.1	min.	Beta
Po^{218}	b	-	3.05	min.	Alpha
Po^{214}	b	-	1.6×10^{-4}	sec.	Alpha
Bi^{214}	b	-	19.7	min	Beta
Pb^{214}	b	-	26.8	min.	Beta
Rn^{222}	(gas)	Lung	3.8	days	Alpha

[a] MPC_w value is the maximum permissible concentration in water, for average member of the general population. [b] No value given.

*From International Commission on Radiological Protection, (ICRP).

These values are given by the International Commission on Radiological Protection (ICRP) and the National Committee on Radiation Protection (NCRP). Radium^{226}, with the lowest MPC of all radioisotopes, is the radiological contaminant of greatest concern in uranium mill wastes discharges. For mixtures about which nothing is known or about which it is known only that radium is present, the MPC is 10 μμc/l. From studies it appears that all but 1 or 2 per cent of the radium originally present in the ore is contained in the waste tailings. This undissolved radium will be leached from these solids to overlying waters and represents a highly significant source of very long-term contamination of receiving waters.

In streams and lakes receiving radioactive wastes, an uptake of the activity by aquatic organisms can generally be observed. These organisms provide a good indication of past contamination since they retain the activity taken up during prior periods. Fish being highly mobile present a relatively erratic pattern of contamination. Attached algae and aquatic insects, on the other hand, reflect contamination history at a specific place and, therefore, provide a good picture of past contamination.

BACKGROUND RADIOACTIVITY

No information is available on the naturally-occurring levels of radioactivity present in ground and surface waters, lake muds, or aquatic biota prior to the start-up of mining operations, in the Elliot Lake area. In a drainage basin such as the Serpent River the background radioactivity will be variable, as water coming into contact with ore deposits rich enough to mine economically will leach out higher concentrations of radioisotopes than waters contacting mineral deposits deficient in uranium, thorium, radium, etc.

With the advent of nuclear weapons and their testing, a second source of "background" radioactivity, namely the fallout from these tests, has been added and as a result the gross beta background radioactivity of surface waters is now somewhat higher than it was a decade ago. This increase is slight, and usually the background content of surface waters will be well below the MPC for gross radioactivity content of 100 uuc/ litre. (NCRP recommendation).

The following tables show some limited data on background radioactivity levels in lakes unaffected by uranium mill wastes in the Serpent River watershed. In the discussion to follow which will deal primarily with the results of biological samples from three lakes (Dunlop, Quirke

and Pecors), Dunlop Lake will be considered the control or reference lake as it has not received any mill wastes.

BACKGROUND QUALITY OF LAKE WATERS

UNAFFECTED BY URANIUM MILL WASTES

Radioactivity μμc/litre

Lake	Gross Alpha	Gross Beta	Radium Content
Benner Lake	3	21	0.1
Quellette Lake	<1	<1	0.1
Popeye Lake	5	22	0.1
Sheriff Lake	2	28	0.7
Teasdale Lake	1	8	0.1

NOTE: Above samples collected January 20, 1964

Dunlop Lake	0.5
Little Quirke Lake	0.1
Quellette Lake	0.2
Sheriff Lake	0.5
Ryan Lake	<0.1

NOTE: Above samples collected October 16--18, 1961.

Table III is a summary of the radium concentration found in the mill effluents from the four mines still operating in the Elliot Lake area and discharging to the Serpent River watershed. These values cover the sampling period January to November, 1963, and were obtained by averaging 11 monthly composite samples:

T A B L E III

Radium Content μμc/litre

Mine	Mill Tailings Discharge			Tailings Area Discharge		
	Avg.	Max.	Min.	Avg.	Max.	Min.
Denison	250	417	40	187	535	64
Stanrock	299	460	162	206	380	99
Milliken	720	925	570	-	-	-
Algom Nordic	564	867	390	217	287	135

SIGNIFICANCE OF BIOLOGICAL AND ENVIRONMENTAL SAMPLES

As we know, organic pollutants undergo biochemical oxidation resulting from bacterial attack in a stream, and thereby are destroyed generally within a few days after they are discharged. In this sense, the stream has a self-purification capacity for organic wastes. No similar process occurs for radioactive materials in the stream environment. In flowing downstream, the concentration of a specific radioisotope will be reduced by dilution with additional uncontaminated tributary flow, and by natural radioactive decay according to a fixed and immutable half-life for that particular radioisotope. Dilution, of course, reduces the water concentration but not the total amount of radioactive material present. Both the concentration and quantity of material present may be apparently reduced as the result of sedimentation of suspended solids and uptake by aquatic organisms. Neither of these actions, however, reduces the total amount of radioisotope present in the stream environment; the material is simply transferred to other phases of the environment, namely, bottom deposits or biological cell content. Stream self-purification in the accepted sense does not occur, and radioactive decay according to fixed half-life is the only means by which the total amount of radio-

activity in the stream environment is diminished.

Water Samples

Samples of river water are one index of the degree of radioactive contamination of a stream and its effects in terms of water use. This type of sample can yield data as to the gross radioactivity content of the flowing water, in both dissolved and suspended form, and if desired, water samples can be analyzed for specific radioelements such as radium. The interpretation and significance of results thus obtained is dependent upon how representative a sample has been collected.

Biological Samples

A second and important index of the extent of radioactive contamination of streams is obtained by assay of samples of the stream biota. During their growth, living organisms, including all of the stream biota, take in and metabolize a wide variety of chemical elements. The extent of uptake varies greatly for different elements, depending upon such factors as availability, chemical form, the needs of the organism, and environmental factors. As a result, the biota can be expected to constitute at least a partially selective sampling device, concentrating certain radioelements to high degree and essentially rejecting others. However, this selectivity cannot be expected to recur in just the same way from stream to stream, due to differences in waste characteristics, and differences in the stream environment.

The radioactivity measured in connection with samples of stream biota may represent either absorbed or adsorbed material. If the former, it is indicative of dissolved or water-soluble radioelements. If it is adsorbed material, it may represent partly insoluble radioelements actually outside the cell walls. Present methods of analysis do not generally permit making a clear distinction here. In either case, if thorough cleaning

(e.g. washing) of the sample does not reduce its radioactivity, then the remaining material becomes an integral part of the life cycle of the stream biota.

Depending upon the recent temperature, stream flows, etc., a biological sample is cumulative, and represents a considerable period of time, whereas, a grab sample of water, represents the condition only at the instant of sampling. Filamentous algae attached to rocks, for example, may have been growing at that stream location for weeks or even months, continuously absorbing nutrients from the water flowing by. Bottom animals, similarly, will have been present for weeks, if they are found at all. While fish are migratory, the range of many species is relatively limited and they, too, can represent exposure of long duration in a limited stream reach. A grab sample of stream biota is therefore really a cumulative sample, and hence very useful at stream locations below sources of radioactive contamination. Because the organisms concentrate many of the radioelements to some degree or other, biological samples are of special value where it is desired to identify specific radioisotopes. In that event, they provide a relatively concentrated sample of small bulk.

Mud Samples

The third type of sample indicative of stream contamination by radioactivity is the river mud or silt. Radioelements from an upstream source may be deposited on the river bed in two major ways: insoluble materials will settle out in reaches of sufficiently low stream velocity; or, as the result of chemical and biochemical alteration of waste constituents, they may be coagulated and settled upon mixing with the stream water. Hence, this type of deposited material is different in nature from the radioactivity found with the stream biota, and, depending on the character of the wastes, may include radioisotopes essentially absent from the

biological samples.

Depending upon the recent flow history of the stream (i.e. droughts and flood flows) and its hydraulic characteristics, mud or silt samples may reflect weeks or even months of accumulation of settleable radioactive material. The bottom deposit samples are therefore also cumulative as compared to the instantaneous nature of grab samples of water. On the other hand, if representative water samples are analyzed for both dissolved and suspended radioactivity, they can provide data for an estimate of the probable amount of radioactivity that will settle.

The studies of stream contamination by uranium mills elsewhere provide an excellent example of the importance of river mud samples. Such samples, taken for a distance of 40 miles below one such ore refinery, indicated significant alpha radioactivity increases at all sampling points. There is little doubt that waste solids from this mill could be tracked a considerably greater distance by means of mud samples. In this way it is possible to form some estimate not only of existing levels of contamination, but also of the rate of build-up of contamination during the history of the operation of the mill.

As has been noted, biological and mud samples are cumulative in nature; whereas, grab samples of river water are instantaneous. Infrequent grab samples of water, taken alone, are not adequate for purposes of radioactive waste control. However, if automatic water sampling equipment is not available, grab samples of water, taken in conjunction with adequate biological and mud sampling, can provide sufficient data for adequate interpretation in terms of stream conditions.

CONDUCT AND ORGANIZATION OF BIOLOGICAL SURVEY

The survey undertaken was of a preliminary nature. The purpose was to collect samples of representative forms of life so that a comparison of radioactivity could be made between specimens taken from lakes which received wastes and those which did not. This data would supply general information on possible hazards associated with use and would provide the basis for designing a more sophisticated study, should this be indicated.

The lakes selected for sample collection were Dunlop, Quirke and Pecors. Dunlop Lake receives no wastes and may be considered as a control. Quirke Lake was known to be the most heavily contaminated and was chosen for this reason. Pecors Lake receives the flow from the Quirke Lake drainage area, and also receives diluted wastes from a group of mines south of Quirke Lake. Pecors Lake may be considered as being representative of the waters presently entering the main stem of the Serpent River.

It was planned that representative samples of the following would be taken from each lake; algae (attached), plankton, crustacea, aquatic insects, clams, fish, bottom mud, and lake water. Figure II indicates the collecting locations in each lake sampled. Table IV indicates the actual samples collected from each lake and the method used for procuring them.

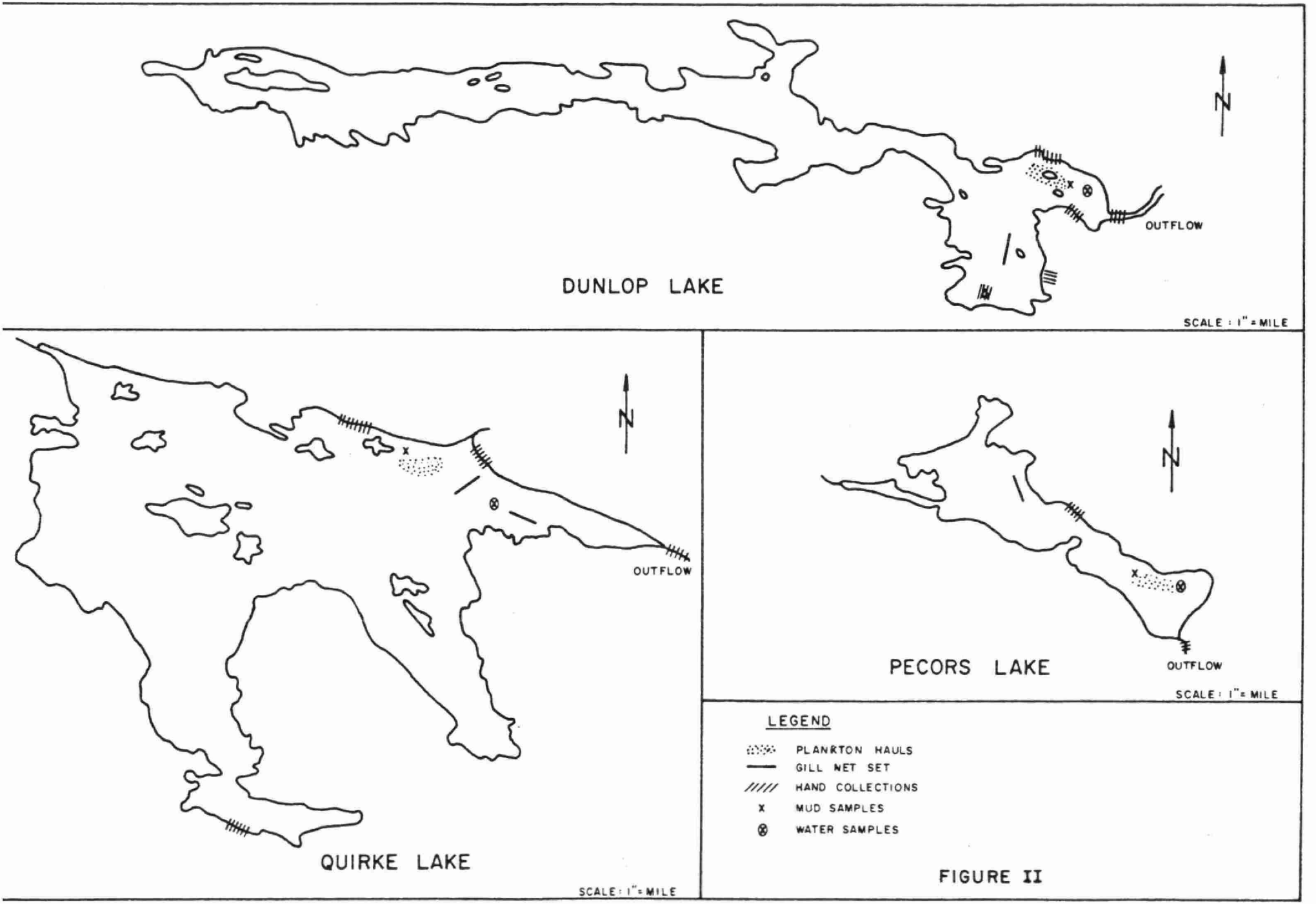

OUTFLOW
DUNLOP LAKE
SCALE : 1" = MILE
OUTFLOW
QUIRKE LAKE
SCALE : 1" = MILE
PECORS LAKE
OUTFLOW
SCALE : 1" = MILE
LEGEND
PLANKTON HAULS
GILL NET SET
HAND COLLECTIONS
MUD SAMPLES
WATER SAMPLES

FIGURE II

T A B L E IV

Samples	Collection Method	Dunlop Lake	Quirke Lake	Pecors Lake
Plankton	50 minute tows, #10 and #20 nylon nets	X	X	X
Algae (attached)	Hand collection		X	X
Crustacea (Crayfish)	Hand collection	X	X	X
Aquatic insects	Hand collection	X		X
Clams	Hand collection			X
Fish - Whitefish	Gill nets	X	X	X
- Cisco	Test set			
- Pickerel		X	X	X
- Others				

The sampling of life in the lakes was simply a qualitative collection of as many groups as possible in the limited time available for collecting. The waters in the area are normally of low productivity. Collections indicated Quirke and Pecors Lakes to be considerably less productive than Dunlop Lake and in some instances no sample or only small samples of the representative groups were available. Samples of only plankton, crayfish, and fish were collected from all three lakes.

Each group of organisms was held separately in plastic bags, frozen at the end of each day and held in this condition until submitted to the Ontario Department of Health for analyses.

Gross alpha and beta activities of water, muds, and biological material were measured at the Radiation Protection Laboratory of the Ontario Department of Health. Briefly water samples were filtered, the filtered solids were dried, weighed, and counted. Gross activity of mud

samples was obtained by counting a weighed, dried sample. The gross activity of biological materials was obtained by counting a weighed sample of ash. Results for biological samples are reported in terms of both ashed weight and blotted dry weight. It should be noted that ashed weights are considered to provide more accurate and reproducible results, due to the factor of self-absorption inherent in bulk samples.

DISCUSSION OF RESULTS

Water Analyses

Table V shows the results of the lake water samples collected during the survey. For reference purposes, and as previously stated under the section of this report titled "Background Radioactivity," Dunlop Lake results will be considered as representing natural background activity.

The soluble radium results of 0.2 μμc/l for Dunlop Lake is quite consistent with the results obtained on other samples taken from this source on previous occasions. The radium results for Quirke Lake and Pecors Lake are respectively 40 μμc/l and 26 μμc/l, and both of these lakes are in the chain of receiving waters for tailing area discharges.

T A B L E V

Quality of Lake Waters

Lake	Radioactivity μμc/litre		
	Gross Alpha	Gross Beta	Radium
Dunlop	1	17	0.2
Quirke	220	130	40
Pecors	97	82	26

TABLE V continued

Tests made on undissolved solids

	Gross Activity	
	Alpha μμc/mgm	Beta μμc/mgm
Dunlop	<1	10
Quirke	81	110
Pecors	13	20

At the peak period of mining activity in the area (1958), Quirke Lake was receiving water wastes from four mines processing an estimated 15,000 tons of ore per day, while Pecors Lake, being located further down the chain of lakes in the watershed, was receiving diluted Quirke Lake water and, in addition, was affected by wastes from four other mines with a total production of 11,500 tons per day which discharged in the chain above Pecors. One would expect the level of soluble radium in Pecors to be less than that of Quirke due to dilution.

Mud Samples

Table VI indicates the results of analyses of lake mud samples. The Dunlop Lake sample is much lower in total radium content than either Quirke or Pecors. The gross alpha and beta counts are significantly higher in both Quirke and Pecors Lakes.

T A B L E VI

QUALITY OF LAKE MUDS

	Radioactivity μμc/gm dry weight			Uranium
Lake	Gross Alpha	Gross Beta	Total Radium	μgm/gm dry wt.
Dunlop	68	88	1.2	5
Quirke	340	220	15	95
Pecors	270	160	13	20

Samples of bottom mud taken from radioactive analyses were collected with an eckman dredge. This is a clam like device which removes a sample of the bottom of varying depth depending on the nature of the substrate. A thin layer of soil at the surface is likely to be the most heavily contaminated and while an attempt was made to collect the top inch, the samples were probably not exactly comparable. Other more sophisticated procedures are available that would permit quantitative comparisons.

Biological Samples

Table VII indicates the radioactivity content of biological samples. Plankton from both Quirke and Pecors Lakes had gross alpha activities far in excess of the average gross alpha activity of 90 $\mu\mu$c/gm (ashed weight) found for three samples in Dunlop Lake. Plankton in Quirke Lake at 12,900 $\mu\mu$c/gm and Pecors at 3,140 $\mu\mu$c/gm (average) exhibited alpha activity 140 and 35 times, respectively, above the level of activity found in Dunlop Lake.

Filamentous algae samples were obtained from only Quirke and Pecors Lakes, but the magnitude of gross alpha radioactivity of 17,600 $\mu\mu$c/gm for Quirke and 750 $\mu\mu$c/gm for Pecors is indicative of radiological pollution.

Bottom insects were not readily obtainable from Quirke Lake but a comparison of the level of gross alpha activity found in Dunlop and Pecors for insects indicated that the level was 30 times higher in Pecors than in Dunlop. A similar comparison was noted with crayfish, where sample results show levels of gross alpha activity 40 times greater in Quirke and 30 times greater in Pecors than the background level of 24 $\mu\mu$c/gm found in Dunlop Lake.

Only one sample of clams was taken and this came from Pecors Lake. The analysis of the flesh showed it to have an alpha count of 26,300 $\mu\mu$c/gm ash and a radium concentration of 18 $\mu\mu$c/gm wet weight. This was the highest

Table VII. RESULTS OF RADIOACTIVE ANALYSES

	Gross Count	Dunlop Lake		Quirke Lake		Pecors Lake	
		μμc/gm Ash	μμc/gm Original	μμc/gm Ash	μμc/gm Original	μμc/gm Ash	μμc/gm Original
Algae (attached)	alpha	NC	NC	17600	420	750	71
	beta	NC	NC	1400	33	940	89
Plankton Phyto and Zoo	alpha	90	3.1	12900	22	3140	2.6
	beta	2430	23.1	8250	14	3150	3.24
Cray Fish	alpha	24	2.2	1150	85	620	62
	beta	350	32	900	67	290	29
Aquatic Insects	alpha	50	0.2	NC	NC	1690	59
	beta	805	2.9	NC	NC	270	9.5
Snails	alpha	30	4.3	NC	NC	NC	NC
	beta	475	68	NC	NC	NC	NC
Clam Shell	alpha	NC	NC	NC	NC	435	384
	beta	NC	NC	NC	NC	208	180
Clam Flesh	alpha	NC	NC	NC	NC	26300	330
	beta	NC	NC	NC	NC	11900	150
Fish							
Whitefish - Bone	alpha	<1	<1	4	NA	21	12.5
	beta	530	405	61	NA	23	13.5
- Flesh	alpha	<1	<0.01	8	NA	<1	0.015
	beta	700	10	380	NA	620	8.7
Cisco-Whole Fish	alpha			245	6.8	13	0.5
	beta			520	14.5	250	8.8
Walleye-Whole Fish	alpha	<1.0	<0.03				
	beta	350	13.5				
Trout - Bone	alpha					54	41
	beta					157	119
- Flesh	alpha					14	0.2
	beta					390	5

NC - Not Collected; NA - Not Analysed.

concentration of radio-activity that has been found in any biological or environmental sample.

The samples of fish analysed for radioactivity were from individual specimens and a considerable variation in the results was found. The alpha count on all the Dunlop Lake specimens, however, was less than one μμc/gm ash for both bone and flesh while the average of all determinations in the other lakes was 29 μμc/gm ash. This indicates that while radioactivity has not been accumulated to the same degree by fish as by other organisms, a definite increase has occurred. Further studies using larger numbers of individuals of various species is needed to establish a precise figure. Further information is also necessary to establish the source of the alpha radiation so that any hazard associated with the long-term consumption of such fish can be evaluated by competent health authorities.

SUMMARY

The clean water location or reference point of this survey, Dunlop Lake, exhibited a soluble radium content of 0.2 μμc/l which was consistent with the values recorded for other lakes in the area known to be unaffected by wastes from uranium mills. This is to be compared with the levels of 40 and 26 μμc/l found respectively in Quirke and Pecors Lakes. Suppliers of domestic water using Quirke Lake as a source have been advised by this Commission that this water is no longer suitable for domestic purposes, and an alternate source must be obtained.

The radioassay results on selected biological samples indicate a large uptake of gross alpha radioactivity, by the aquatic biota in Quirke and Pecors Lakes. The maximum gross alpha activity in any of the aquatic biological samples obtained from Dunlop Lake was 112 μμc/gm (ashed weight).

This is to be compared with such values as 17,600 μμc/gm (ashed weight) for a sample of algae from Quirke Lake and 26,300 μμc/gm (ashed weight) for a sample of clam flesh obtained from Pecors Lake. This single sample of clams showed such high activity that it was tested for radium and a concentration of 18,000 μμc/kg of wet flesh was found. The allowable concentration of radium in foods is 3 μμc/kilogram of live material. It is likely that most of the alpha radioactivity found in biological samples came from radium but this was only confirmed in the clam flesh. The silt or mud samples, taken from Quirke and Pecors Lakes were approximately 12 times higher in radium than the sample taken from Dunlop Lake.

To a great extent pollution of Quirke and Pecors Lakes by suspended material has been minimal from the mines discharging to these lakes because of good tailings control in the basins and lakes used for disposal. The neutralization of the acid mill wastes by the mines in the Elliot Lake area, in addition to eliminating the quite significant harmful effects of excess acidity on the aquatic life of receiving waters, reduces the solubility of radioactive and chemical pollutants by causing their precipitation and subsequent retention in the waste ponds. However, this reduction of dissolved radium does not appear to have prevented a build-up of radium from taking place in the receiving waters affected by tailings area discharges.

The magnitude and distribution of radioactive pollution in the Serpent River watershed cannot be fully evaluated from the existing samples as sampling was confined to only two lakes directly affected by mill wastes. However, enough evidence has been gathered to indicate that the observed radioactivity is an important environmental contaminant.

RECOMMENDATIONS

At present insufficient information is available on the distribution and intensity of radioactive substances within the Serpent River watershed and on the effects of chemical changes in the water on their productivity.

It would appear that in order to minimize further pollution, a careful assessment must be made of present refining processes and waste treatment and disposal practices. Proper design and adequate maintenance of existing tailings areas is of particular importance in this regard.

For these reasons an intensive survey of the watershed is recommended which should include at least the following considerations:

1) An increased sampling program in all waters affected by uranium mining wastes to monitor present and future changes in radioactivity and water chemistry.
2) A study of the level of radioactivity in the biota from representative waters from above the source of wastes to the North Channel of Lake Huron.
3) An identification of the isotopes responsible for emissions in both biological and environmental samples.
4) A review of all radiological findings by competent authorities to assess whether any hazard is associated with the use of the waters or the life they contain.
5) A study of the changes in the biological productivity of the water to determine quantitatively what changes have taken place and to make recommendations for corrections if necessary.
6) Determine what measures are necessary to further reduce the quantity of pollutants gaining access to the watershed and to impose such measures as are indicated upon all operating and non-operating works.

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DUNLOP LAKE SAMPLES - TAKEN BY ONTARIO WATER RESOURCES COMMISSION - OCTOBER 3--4, 1963 RPL-63-145

Type of Sample	Wt. or Vol. of Sample Used	Wt. of Dry Solids	Drying Temp. °C	% Ash	Ashing Temp. °C	pc/gm Ash alpha	pc/gm Ash beta	pc/gm Original alpha	pc/gm Original beta	pc/Litre alpha	pc/Litre beta	Count Made on	pc Radium	u gm Uranium	Notes
Phyto and Zoo Plankton															
Bottle 1--Plankton	59 gm wet	3.8 gm	40°	0.34	450	112	5600	0.4	19			Ash			
Ambient Fluid	100 ml			0.10	400	7.7	870			8	910	SO_4 Ash			
Bottle 2--Plankton	60 gm wet	1.7 gm	40°	0.28	450	62	1160	0.2	3.2			Ash			
Ambient Fluid	100 ml			0.09	400	15	1150			14	1040	SO_4 Ash			
Bottle 3--Plankton	4.0 gm dry		40°	9.0	450	97	530	8.8	48			Ash			
Ambient Fluid	100 ml			0.088	400	17	1150			15	1010	SO_4 Ash			
Snails	1.6 gm wet			14.4	450	30	475	4.3	68			Ash			Whole Snails Used
Insects	27.9 gm wet			0.36	450	50	805	0.2	2.9			Ash			
Crayfish	57.5 gm wet			9.2	450	24	350	2.2	32			Ash			
White Fish--Bone	8.17 gm dry		80°	76.	450	<1	530	<1	405			Ash			
White Fish--Flesh	579 gm wet			1.44	450	<1	700	<0.01	10			Ash			
Pickerel	42 gm wet			3.86	450	<1	350	<0.03	13.5			Ash			Whole Fish Used
Lake Water	500 ml	160 mgm	80°							1	17	Dried Solids	0.2/ Litre	<10/ Litre	
-Undissolved Solids Ex1000	ml	0.04 "	80°					<1/mgm	10/mgm			Dried Solids			
Lake Bottom Mud	5 gm dry		80°					68/dry gm	88/dry gm			Dried Solids	1.2/ dry gm	5/ dry gm	

Analysed by Radiation Protection Laboratory.

PECORS LAKE SAMPLES - TAKEN BY ONTARIO WATER RESOURCES COMMISSION - OCTOBER 3--4, 1963 RPL-63-146

Type of Sample	Wt. or Vol. of Sample Used	Wt. of Dry Solids	Drying Temp. °C	% Ash	Ashing Temp. °C	pc/gm Ash alpha	pc/gm Ash beta	pc/gm Original alpha	pc/gm Original beta	pc/Litre alpha	pc/Litre beta	Count Made on	pc Radium	u gm Uranium	Notes
Phyto and Zoo Plankton															
Bottle 1--Plankton	156 gm wet	3.9 gm	40°	0.04	450	3730	2700	1.5	1.08			Ash			
Ambient Fluid	100 ml			0.037	400	25	180			9.3	67	SO_4 Ash			
Bottle 2--Plankton	97 gm wet	1.7 gm	40°	0.15	450	2550	3600	3.8	5.4			Ash			
Ambient Fluid	100 ml			0.11	400	17	72			19	79	SO_4 Ash			
Insects	26.9gm wet	7.7 gm	40°	3.5	450	1690	270	59	9.5			Ash			
Crayfish	71.3gm wet	16 gm	40°	10.1	450	620	290	62	29			Ash			
Algae	41.3gm wet	6.9 gm	40°	9.5	450	750	940	71	89			Ash			
Clam--Shell	36 gm dry	36.6 gm		86	450	435	208	384	180			Ash			
Clam--Flesh	65.7gm wet	3.1 gm	40°	1.27	450	26300	11900	330	150			Ash	18/wet gm		
Fish--Sucker--Bone	9.8gm dry		80°	67.5	450	13	25	8.8	17			Ash			
Fish--Sucker--Flesh	230 gm wet			1.32	450	8	390	0.1	5.2			Ash			
White Fish--Bone	7.1gm dry		80°	59	450	21	23	12.5	13.5			Ash			
White Fish--Flesh	330 gm wet			1.4	450	<1	620	<0.015	8.7			Ash			
Pickerel--Bone	3.9gm dry		80°	77	450	22	30	17	23			Ash			
Pickerel--Flesh	254 gm wet			1.4	450	4	360	0.06	5			Ash			Whole Fish Ashed
Cisco	220 gm wet			3.5	450	13	250	0.5	8.8			Ash			
Trout--Bone	5.8gm dry		80°	75.5	450	54	157	41	119			Ash			
Trout--Flesh	406 gm wet			1.3	450	14	390	0.2	5			Ash			
Lake Water	500 ml Ex	150 mgm	80°							97	82	Dried Solids	26/ Litre	<10/ Litre	
-Undissolved Solids	1000 ml	0.3 mgm	80°					13/ mgm dry	20/ mgm dry			Dried Solids			
Lake Bottom Mud	5 gm dry		80°					270/ gm	160/ gm			Dried Solids	13/ dry gm	20/ dry gm	

Analysed by Radiation Protection Laboratory.

QUIRKE LAKE SAMPLES - TAKEN BY ONTARIO WATER RESOURCES COMMISSION - OCTOBER 3--4, 1963 RPL-63-147

Type of Sample	Wt. or Vol. of Sample Used	Wt. of Dry Solids	Drying Temp. °C	% Ash	Ashing Temp. °C	pc/gm Ash alpha	pc/gm Ash beta	pc/gm Original alpha	pc/gm Original beta	pc/Litre alpha	pc/Litre beta	Count Made on	pc Radium	u gm Uranium	Notes
Phyto and Zoo Plankton															
Bottle 1--Plankton	38 gm wet	1.5 gm	40°	0.17	450	12900	8250	22	14			Ash			
Ambient Fluid	100 ml			0.052	400	4.6	72			2.4	38	SO_4 Ash			
Crayfish	36 gm wet	7.8 gm	40°	7.45	450	1150	900	85	67			Ash			
Algae	66 gm wet	18.5 gm	40°	2.4	450	17600	1400	420	33			Ash			
Fish--Cisco	190 gm wet			2.8	450	245	520	6.8	14.5			Ash			Whole Fish Ashed
White Fish--Bone	Not Recorded				450	5	92					Ash			
--Flesh					450	9	340					Ash			
--Bone					450	3	30					Ash			
--Flesh					450	7	420					Ash			
Aquatic Plants	74 gm wet	11.3 gm		3.8	450	4620	340	175	13			Ash			
Lake Water	500 ml	130 mgm	80°							220	130	Dried Solids	40/ Litre	23/ Litre	
-Undissolved Solids	Ex1000 ml	0.02 mgm	80°					81/ mgm	110/ mgm			Dried Solids			
Lake Bottom Mud	5 gm dry		80°					340/ Dry gm	220/ Dry gm			Dried Solids	15/ Dry gm	95/ Dry gm	

Analysed by Radiation Protection Laboratory